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Laser diode optically pumped caesium beam

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Abstract. — This paper presents a theoretical analysis of a possible scheme to realize a strong population difference between the two ground state sub-levels \( F = 4, \, M_F = 0 \) and \( F = 3, \, M_F = 0 \) of caesium atom. Two laser-diode light beams, \( \pi \) polarized, are used to illuminate at right angle an atomic beam. The efficiency of the population difference build up is evaluated for two limiting cases according to the light spectral distribution: the monochromatic and broadband excitations. This analysis takes into account hyperfine and Zeeman structures of the energy levels. Numerical calculations show that the monochromatic excitation leads to steady state population difference close to the value obtained with a single pumping laser; however all the atoms can be brought in the upper level if the two laser lights are considered broadband and uncorrelated. Guidelines are derived to obtain an efficient optically pumped caesium beam frequency standard.

1. Introduction.

Actual caesium beam frequency standards [1] use a deflecting magnet to select a class of atoms having a limited range of velocities and an appropriate hyperfine energy state. The beam is directed into a Ramsey type microwave cavity [2] where the \( F = 4, \, M_F = 0 \rightarrow F = 3, \, M_F = 0 \) transition is induced (clock transition). The resonance is observed with a second deflecting magnet and a hot wire detector. The performance of this type of device is mainly limited by the cavity phase dependent frequency shift and the evaluation of the beam velocity [3].

Arditi and Picqué [4] have shown that it is possible to replace the state selecting magnet scheme by an optical pumping cycle to create the desired population difference; such a system yield higher beam intensity and better efficiency since all the atoms can be used in the pumping cycle. Recently, the advent of simple solid state laser diodes, operating at room temperature and at the appropriate wavelength (852.1 nm), has increased the interest for this proposal [5]. A scheme using two laser diodes in the optical pumping region [6], should transfer all the atoms onto a single sub-level. Therefore, the signal to noise ratio would be increased leading to a more stable frequency standard. Moreover the atomic velocity distribution should be known more precisely thus allowing a better evaluation of the residual frequency shifts limiting the accuracy of the device as a primary frequency standard [7].

In this paper, we present a theoretical analysis of the population difference build up for a caesium beam pumped by two lasers. We compare the results derived with a direct phenomenological approach.
(rate equations) [8] to those obtained with the master equation approach (density matrix) [9]. For each approach, both the monochromatic and broadband excitation are considered. Numerical solutions are computed for each case and application to the realization of a practical frequency standard is discussed.

2. Proposed optical pumping scheme.

Figure 1 shows the scheme of an optically pumped frequency standard; the population inversion is achieved by two pumping lasers and the resonance phenomenon (Ramsey fringes) is detected through fluorescence light induced by a third laser diode. A weak magnetic field B, removes the Zeeman degeneracy. In this work, we limit our attention to the evaluation of the population unbalance created in the optical pumping region. Figure 2 shows the energy levels of the D₂ line and the optical transitions involved in this study: a first laser diode, LD₁, is tuned to the transition between levels g(F = 3) and e(F' = 4) while the second laser, LD₂, is tuned to the transition between levels f(F = 4) and e (Fig. 2b).

If we look up at the table of probability transition coefficients (appendix 1), we see that the transition between the Zeeman sub-levels e(0) ↔ f(0) is forbidden, so if we use π polarized laser light, LD₁ brings the atoms into the level f while LD₂ depopulates the sub-levels f(M) with M ≠ 0; consequently all the atoms are finally accumulated in the sub-level f(0).

The characteristics of the pumping lights are typical of commercially available laser diodes: intensity of 5 mW and spectral width of 30 MHz. Such a spectral width is wider than the natural width of the D₂ transition (5 MHz), but much narrower laser linewidths are possible [10], so the calculation have to be done for broadband and monochromatic excitations.

3. Evolution of energy level population: phenomenological approach.

The phenomenological approach considers the rate equations describing solely the population evolution of the various levels involved in the pumping process. From the Zeeman structure shown on figure 2c, we find that the rate equations for the population n of a given energy state form a set of 25 equations like,

$$\frac{dn}{dt} = \frac{d^{(1)}n}{dt} + \frac{d^{(2)}n}{dt} + \frac{d^{(3)}n}{dt}. \quad (1)$$

The first and second terms of the right hand side are respectively due to absorption and stimulated emission processes. The third term expresses the spontaneous emission process.

We can write directly the evolution of the population of any Zeeman sub-level of the excited state nₑ and

Fig. 2. — Energy level diagram corresponding to the D₂ line of caesium atom. a) Fine and hyperfine structure; b) Laser excitations; c) Zeeman structure of the levels e, f, and g. The energy gaps are not to scale.
ground states $n_f, n_g$ as:

$$
\frac{dn_e}{dt} = W_{f \rightarrow e} n_f + W_{g \rightarrow e} n_g - (W_{e \rightarrow f} + W_{e \rightarrow g}) n_e - \left( \sum_f A_{e \rightarrow f} + \sum_g A_{e \rightarrow g} \right) n_e \tag{2}
$$

$$
\frac{dt}{dt} = - W_{f \rightarrow e} n_f + W_{e \rightarrow f} n_e + \sum_f A_{e \rightarrow f} n_e \tag{3}
$$

$$
\frac{dx}{dt} = - W_{g \rightarrow e} n_g + W_{e \rightarrow g} n_e + \sum_g A_{e \rightarrow g} n_e . \tag{4}
$$

The coefficients $A_{e \rightarrow f}$ and $A_{e \rightarrow g}$ are the spontaneous emission rates from a Zeeman sub-level $e$ to a Zeeman sub-level $f$ or $g$.

We have:

$$
A_{e \rightarrow f} = \frac{\alpha_e^2}{\Gamma} \tag{5}
$$

where $\Gamma = 1/\tau$ is the natural width of the D$_2$ transition ($\tau = 30$ ns) and $\alpha_{e \rightarrow f}$ is the transfer coefficient as established in appendix I.

The coefficients $W_{e \rightarrow f}$ and $W_{e \rightarrow g}$ are stimulated emission rates and the coefficients $W_{f \rightarrow e}$ and $W_{g \rightarrow e}$ are the corresponding absorption rates. We recall that $W_{f \rightarrow e} = W_{f \rightarrow e}^*$ and $W_{g \rightarrow e} = W_{g \rightarrow e}^*$. In the case of monochromatic excitation centred on the atomic absorption line, we have:

$$
W_{e \rightarrow f(a)} = \frac{\lambda_i^3}{2 \pi hc} \frac{P_i}{S} \frac{1}{\Gamma} A_{e \rightarrow f(a)} \tag{6}
$$

where $h$ is Planck constant, $c$ is the speed of light in vacuum, $\lambda_i$ and $P_i/S$ represent the wavelength and the intensity of the laser LD$i$ respectively ($i = 1, 2$).

On the other hand, if the light excitation is not monochromatic but is supposed to be distributed over a Lorentzian lineshape of width $\Delta_p$, the $W$ values in equation 6 must be reduced by a factor $\Gamma/\Delta_t$.

In equations 2, 3 and 4, we have neglected the relaxation processes of the ground state.

We postulate that all the 16 Zeeman sub-levels of the ground state are equally populated when the Cs beam enters into pumping region. We suppose that the laser intensities are uniformly distributed all over the possible optical transitions. Finally, the atomic beam is assumed to be monokinetic.

The evolution of the population difference between the two field-independent ground state levels $\Delta n(t) = n_f(t) - n_g(t)$ is shown on figure 3 for several laser intensities. We can see that a complete population inversion, bringing all the atoms of the beam into the state $f(0)$, seems possible if the light intensity is sufficiently strong. A light intensity of 30 mW/cm$^2$ corresponds to a typical value nowadays available. We recall that a pumping scheme with a single laser gives 13% population difference (see dashed line). As shown in the next section, different results are obtained when coherence effects induced by laser fields are taken into account.


4.1 Monochromatic excitation.

4.1.1 Master equation. — In this section we use the density matrix formalism which gives a more rigorous solution. We take a semi-classical model in which the laser field is written classically and the atomic system is treated through quantum-mechanics. The density matrix formalism considers not only the populations but also the coherence created when an interaction couples two levels. The evolution of the density matrix $\rho$, expressed in the Schrödinger picture, obeys the well known equation [11]:

$$
\frac{d\rho}{dt} = \frac{1}{i\hbar} [H, \rho] + R . \tag{7}
$$

The Hamiltonian $H$ is the sum of the unperturbed Hamiltonian $H_0$ and the interaction term $V$. $R$ represents the relaxation processes introduced phenomenologically.

The interaction term is:

$$
V = -D \cdot E \tag{8}
$$

where $D$ is the electric dipole moment and $E$ the total electric field. The lasers are assumed to emit monochromatic plane waves, then:

$$
E_1(t) = \frac{1}{2} E_{1,0}(e^{i\omega_1 t} + e^{-i\omega_1 t}) e_1 \tag{9}
$$

$$
E_2(t) = \frac{1}{2} E_{2,0}(e^{i\omega_2 t} + e^{-i\omega_2 t}) e_2 . \tag{10}
$$

In our case, both lasers are π polarized so $e_1 = e_2 = e_x$.

The interaction term becomes:

$$
V = -\frac{1}{2} D_x [E_{1,0}(e^{i\omega_1 t} + e^{-i\omega_1 t}) + E_{2,0}(e^{i\omega_2 t} + e^{-i\omega_2 t})] . \tag{11}
$$

For each density matrix element $\rho_{ij}$ equation 7 yields:

$$
\frac{d\rho_{ij}}{dt} = -i\omega_{ij} \rho_{ij} + \frac{1}{i\hbar} \sum_k (V_{ik} \rho_{kj} - \rho_{ik} V_{kj}) + R_{ij} . \tag{12}
$$
with \( \omega_{ij} = (\epsilon_i - \epsilon_j)/\hbar \). \( E_{ij} \) is the energy of level \( i(j) \), \( V_{ik} \) is a matrix element of \( \mathbf{V} \) and \( R_{ij} \) is the relaxation term. Diagonal elements are populations, while off-diagonal elements are coherences. Three kind of coherences can be encountered:

- Optical coherences \( \rho_{ee} \), \( \rho_{es} \).
- Hyperfine coherences \( \rho_{ss} \).
- Zeeman coherences \( \rho_{es}, \rho_{ss} \) or \( \rho_{tt} \).

In the interaction representation with respect to \( H_0 \) [11], equation 11 becomes:

\[
\frac{d\tilde{\rho}_{ij}}{dt} = \frac{1}{i\hbar} \sum_k (\tilde{V}_{ik} \tilde{\rho}_{kj} - \tilde{\rho}_{ik} \tilde{V}_{kj}) + \tilde{R}_{ij}. \tag{12}
\]

Within the rotating wave approximation, we obtain

\[
\frac{d\tilde{\rho}_{ij}}{dt} = \frac{iE_{1,0}}{2\hbar} \sum_k [D_{ik} \tilde{\rho}_{kj} e^{i(E_{ik} - \omega_{1,0})t} - \tilde{\rho}_{ik} D_{kj} e^{i(E_{kj} - \omega_{1,0})t}] \\
+ \frac{iE_{2,0}}{2\hbar} \sum_t [D_{it} \tilde{\rho}_{jt} e^{i(E_{it} - \omega_{2,0})t} - \tilde{\rho}_{it} D_{jt} e^{i(E_{jt} - \omega_{2,0})t}] + \tilde{R}_{ij} \tag{13}
\]

where \( \omega_{ee} = +1 \) if \( E_m > E_n \) or \( -1 \) otherwise, and \( D_{ik} = \langle i| D| k \rangle \).

In the summations, only the terms with \( \omega_{mn} - \omega \approx 0 \) (\( i = 1, 2 \) according to the laser) bring a significant contribution, so the sum is restricted to the corresponding \( D_{mn} \) terms. In appendix II we give the explicit writing of equation 13 for populations and coherences.

### 4.1.2 Relaxation terms.

The relaxation process is assumed to come solely from spontaneous emission, it is isotropic and affects both the populations and the coherences. For populations (see equations 2, 3, 4 and references), we can write:

\[
\begin{align*}
\tilde{R}_{ee} & = - \left( \sum_t A_{e,-t} + \sum_k A_{e,-k} \right) \tilde{\rho}_{ee} = -\Gamma \tilde{\rho}_{ee} \tag{14} \\
\tilde{R}_{tt} & = \sum_k A_{k, e} \tilde{\rho}_{ee} \tag{15} \\
\tilde{R}_{ss} & = \sum_k A_{k, e} \tilde{\rho}_{ee} \tag{16}
\end{align*}
\]

The optical coherences relax according to [9]:

\[
\tilde{R}_{ef} = -\left( \Gamma_{e,-f} + \frac{\Delta E_{ef}}{\hbar} \right) \tilde{\rho}_{ef} \tag{17}
\]

with a similar equation for \( \tilde{R}_{ef} \).

The term \( \Delta E_{ef}/\hbar \) is the radiative Lambshift and as usual, it will be included in the atomic resonant frequency. \( \Gamma_{ef} \) is the damping rate of the optical coherence, its value is \( \Gamma_{ef} = \frac{1}{2} \left( \sum_{t} A_{e,-t} + \sum_k A_{k,-e} \right) = \Gamma/2 \). For the ground state, the hyperfine and the Zeeman coherences can be written:

\[
\begin{align*}
\tilde{R}_{gg} & = \sum_{k} A_{k, e} \tilde{\rho}_{ee} \tag{18} \\
\tilde{R}_{ft} & = \sum_{n} A_{n, f} \tilde{\rho}_{ee} \tag{19} \\
\tilde{R}_{gs} & = \sum_{n} A_{n, f} \tilde{\rho}_{ee} \tag{20}
\end{align*}
\]

The transfer coefficients \( a_{ij} \) are defined in appendix I. The \( M_f \) are magnetic quantum numbers. Equation 18 shows that the evolution of a hyperfine coherence \( \tilde{\rho}_{fg} \) with \( M_f = M_g \) depends on the population \( \tilde{\rho}_{ee} \); on the other hand, if \( M_f \neq M_g \) it is linked to a Zeeman coherence \( \tilde{\rho}_{ee} \) which is null in our case since we use \( \pi \) polarized light. For this same reason, \( \tilde{R}_{ft} = \tilde{R}_{gs} = 0 \).

### 4.1.3 Numerical results.

Equation 13 has been solved numerically in various cases. Figure 4 shows the evolution of \( \Delta n(t) = n(t) - n(0) \) for two resonant lasers of equal intensity. The stationary value of \( \Delta n \) is only 14 % instead of 100 % as for the rate equation treatment, it is very close to the result obtained with one laser (13.6 %). A thorough numerical study has shown that this result is verified over a wide range of pumping power especially at very weak power value. If the frequency of one laser is untuned while the other one is kept tuned to the transition, the effect remains about the same although the population inversion can increase slightly.

This relatively weak population unbalance is due to the creation of hyperfine coherences when the laser frequency difference equals the hyperfine splitting. This phenomenon is related to « black line » [13-15], as well as « stimulated resonance Raman transition » [16] or « coherent population trapping » [17] effects.

### 4.2 BROADLINE EXCITATION.

In the previous section the coherence time of the lasers were infinite, it is interesting to know if the hyperfine coherences remain.
still created when the laser frequency spectrum broadens. This situation is more realistic since available laser diodes are far from being monochromatic.

In the following, we adapt the formalism developed by C. Cohen-Tannoudji for broadband excitation [9] to our case (hyperfine structure and two lasers excitation). We assume that the light excitations are broadband with respect to the Zeeman splitting but narrow band compared to the hyperfine splitting.

The power spectrum of each laser light can be written as a Lorentzian line of width $\Delta_i$, centred at $\omega_i$:

$$P(\omega) = P_i g_i(\omega) = \frac{P_i}{\pi} \frac{\Delta_i/2}{(\omega - \omega_i)^2 + (\Delta_i/2)^2}$$

(21)

where $i$ stands for laser 1 or 2, $P_i = \int_{-\infty}^{\infty} P(\omega) \, d\omega$ gives the total power of the laser.

This power spectrum is associated to an electric field $E_i(t) \, e_i$ where $e_i$ is the polarization vector and $E_i(t)$ the instantaneous amplitude. $E_i(t)$ is a random function of time which is postulated stationary [18]. Its autocorrelation function is defined as:

$$R_{E_i}(\tau) = \langle E_i(t) E_i(t - \tau) \rangle$$

(22)

where the bar means a time average.

The spectral density of the field fluctuations is:

$$I_i(\omega) = \frac{1}{2\pi} \frac{P_i}{C \, S} g_i(\omega)$$

(23)

It results that the autocorrelation function can be expressed as:

$$R_{E_i}(\tau) = R_{E_i}(0) \exp\left(-\frac{|\tau|}{\tau_i}\right)$$

(24)

where $\tau_i = 1/\Delta_i$ is the coherence time of the laser wave.

The pumping times $T_{pf}$ and $T_{pg}$ are defined by the relations 9:

$$\frac{1}{T_{pf}} = \frac{2}{h^2} I_2(\omega_{ef}) \left| D_{ef} \right|^2$$

(25)

and

$$\frac{1}{T_{pg}} = \frac{2}{h^2} I_1(\omega_{eq}) \left| D_{eq} \right|^2.$$ 

(26)

The excitation is said « broad line » if $\tau_i \ll T_{pf,pg}$. In this case, the laser fields cannot build up coherences between successive optical transitions. For a laser linewidth of 30 MHz $\tau_i \approx 5$ ns thus the excitation is broad line for a 30 mW/cm$^2$ intensity ($T_p = 76$ ns), however for 300 mW/cm$^2$ $T_p = 7$ ns and the approximation is no longer valid.

Since we deal with two independent laser fields, the interaction term is the sum of two statistically independent random functions with coherence times $\tau_1$ and $\tau_2$.

We write:

$$V(t) = -D \cdot e_1 \, E_1(t) - D \cdot e_2 \, E_2(t).$$

(27)

The substitution of $V(t)$ into equation 12 gives cross-product terms $E_i(t) E_j(t')$ which vanish after time averaging since the field fluctuations are uncorrelated (see appendix III). Also we find that populations are coupled only to populations as it is the case for one broadband laser excitation [9].

In the interaction representation and within the rotating wave approximation, the evolution of the population becomes:

$$\frac{d\rho_{ee}}{dt} = -\left(\frac{1}{T_{pg}} + \frac{1}{T_{pf}}\right)\rho_{ee} + \frac{1}{T_{pf}} \rho_{ft} + \frac{1}{T_{pg}} \rho_{ss} + \bar{\rho}_{ee}$$

(28)

$$\frac{d\rho_{ft}}{dt} = \frac{1}{T_{pf}} \rho_{ee} - \frac{1}{T_{pf}} \rho_{ft} + \bar{\rho}_{ft}$$

(29)

$$\frac{d\rho_{ss}}{dt} = \frac{1}{T_{pg}} \rho_{ee} - \frac{1}{T_{pg}} \rho_{ss} + \bar{\rho}_{ss}$$

(30)

where the terms $\bar{\rho}_{ee}$, $\bar{\rho}_{ft}$ and $\bar{\rho}_{ss}$ are expressed by equations 14, 15 and 16 respectively.

The population difference build up $\Delta n(t)$, calculated from equations 28, 20 and 30 is shown on figure 5 for various laser power.

We see that a full population inversion (100 %) is achievable if the duration of the interaction is sufficiently long. The required time to reach a value close to 100 % is shortened if the laser power increases. These results are similar to those obtained through phenomenological approach in the broadband case (Curves 3 and 5 are not identical because the laser spectral power distributions are different compared to the atomic absorption line). This fact holds only because the two laser lights are broadband and uncorrelated. Dalton and Knight [17, 23] showed that, if the field fluctuations are cross-correlated, $\Delta n$ should decrease because the coherence effect acts again.

Fig. 5. — Time evolution of the population difference between levels $F = 4, M_F = 0$ and $F = 3, M_F = 0$ calculated with the density matrix model for broadband excitations.
5. Sequential pumping.

If the laser lights are narrow band or if the intensities are sufficiently strong, the pumping time may become shorter than the correlation time of each laser field. In this case the resulting population difference lies somewhere between 14% and 100%. However, a sequential pumping scheme where the light intensities are chopped alternatively may ensure a full pumping efficiency. Using equation 13 where the electrical fields are turned on accordingly to the sequence indicated on figure 6, we obtain the population difference, $\Delta n$, shown on this figure. The turn on time must be longer than the radiative lifetime of the excited state in order to realize efficient pumping. Otherwise the hyperfine coherences created because a field $E_i$ is present while the coherence relative to the other resonance frequency has not yet relaxed (see eq. II.6), prevent complete population inversion. Numerical results show that the lower limit to the turn on time is roughly independent of laser intensity.

6. Conclusion.

This study shows that efficient optical pumping of a Cs beam can be realized with two lasers if they are broadband and uncorrelated.

The spread of the light spectrum should not be too wide, otherwise the hyperfine structure of the excited state will have to be considered. The light intensity should be maintained at a level that ensures a pumping time longer than the correlation time of each laser light. The population inversion would then be of 100% after a few micro-seconds of interaction time in usual experimental conditions. Obviously, each laser must be long term stabilized.

With a narrow laser spectrum, a sequential pumping scheme where the turn on time is longer than the radiative lifetime of the excited state should also give a very efficient pumping.

The model presented here is adequate to evaluate the performance of other possible pumping schemes where $\sigma$ and $\pi$ polarized light and various hyperfine levels of the excited state [19, 20] are involved.

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Appendix I.

Transfer coefficients and transition probabilities. — The spontaneous emission probability between the states $|J_e IF_e M_e \rangle$ and $|J_g IF_g M_g \rangle$ is given by (S.I. units) :

$$A_{F_e M_e \rightarrow F_g M_g} = \frac{\alpha_{\text{sp}}^2}{3 \pi \epsilon_0 \hbar c^2} |\langle J_e IF_e M_e | D | J_g IF_g M_g \rangle|^2 .$$

(1.1)

The standard component $D_q (q = -1, 0, +1)$ of the electric dipole moment $D = e \mathbf{r}$ between the states $|J_e IF_e M_e \rangle$ and $|J_g IF_g M_g \rangle$ may be written as :

$$\langle J e IF e M e | D_q | J g IF g M g \rangle = (-1)^{1+J_e+J_g+M_e+M_g} (2 J_e + 1)^{1/2} \times$$

$$\times (2 M_e + 1)^{1/2} \left( \begin{array}{ccc} F_e & 1 & F_g \\ -M_e & q & M_g \end{array} \right) \left( \begin{array}{ccc} 1 & 1 & F_g \\ 1 & J_g & 1 \end{array} \right) \langle J_e \parallel D \parallel J_g \rangle$$

$$= (-1)^q \langle J e IF e M e | D_{-q} | J g IF g M g \rangle$$

(1.2)

where ( ) and { } are 3-j and 6-j symbols, $\langle J_e \parallel D \parallel J_g \rangle$ is the reduced matrix element of $D$ [21].
If the Zeeman structure is not resolved we must sum over the final sublevels $M_g$ and take the average over the initial sublevels $M_e$:

$$A_{F_e-F_g} = \frac{\omega_{eg}^2}{3 \pi \varepsilon_0 \hbar c} \frac{1}{2 F_e + 1} \sum_{M_e M_g} (2 F_e + 1) (2 F_g + 1) \left( \begin{array}{c c c}
F_e & 1 & F_g \\
-M_e & q & M_g
\end{array} \right)^2 \times
\left( \begin{array}{c c c}
F_e & 1 & F_g \\
J_g & I & J_e
\end{array} \right)^2 \left| \langle J_e \parallel D \parallel J_g \rangle \right|^2. \tag{I.3}
$$

Using the sum rules on 3-\textit{j} symbols, we obtain:

$$A_{F_e-F_g} = \frac{\omega_{eg}^2}{3 \pi \varepsilon_0 \hbar c} (2 F_g + 1) \left( \begin{array}{c c c}
F_e & 1 & F_g \\
J_g & I & J_e
\end{array} \right)^2 \left| \langle J_e \parallel D \parallel J_g \rangle \right|^2. \tag{I.4}
$$

Now if the hyperfine structure are not resolved we must sum over the final levels $F_g$ and take the average over the initial levels $F_e$. Finally the sum rule on 6-\textit{j} symbols gives:

$$A_{F_e-J_e} = \frac{\omega_{eg}^3}{3 \pi \varepsilon_0 \hbar c^3} \frac{1}{2 J_e + 1} \left| \langle J_e \parallel D \parallel J_g \rangle \right|^2 = \frac{1}{\tau}, \tag{I.5}
$$

where $\tau$ is the lifetime of the $J_e$ level. $\tau$ is known from experiment. In the Cs case for the $6 \ ^2P_{3/2}$ level, $\tau$ is 30 ns [22].

We define the transfer coefficients $a_{F_eM_e-F_gM_g}$ by the relation:

$$a_{F_eM_e-F_gM_g} = (-1)^{I_e+J_e+F_e+F_g-M_e} (2 F_e + 1)^{1/2} (2 F_g + 1)^{1/2} \times
\left( \begin{array}{c c c}
F_e & 1 & F_g \\
-M_e & q & M_g
\end{array} \right) \left( \begin{array}{c c c}
F_e & 1 & F_g \\
J_g & I & J_e
\end{array} \right) (2 J_e + 1)^{1/2}. \tag{I.6}
$$

The values of those coefficients for the transitions $F' = 4 \leftrightarrow F = 4$ and $F' = 4 \leftrightarrow F = 3$ are given in figure 7. They are related to the probability transition by:

$$A_{F_eM_e-F_gM_g} = (a_{F_eM_e-F_gM_g})^2 \frac{1}{\tau} \tag{I.7}
$$

Where we have used the relation:

$$\langle J_e | D_e | M_g \rangle \langle J_e | D_g | M_g \rangle = a_{F_eM_e-F_gM_g} \left( \frac{3 \pi \varepsilon_0 \hbar c^3}{\omega_{eg}^3} \right)^{1/2}. \tag{I.8}
$$

Appendix II.

Explicit form of density matrix element evolution. — Equation 13 gives for the time evolution of the populations:

$$\frac{d}{dt} \tilde{\rho}_{ee} = \frac{iE_1}{\hbar} [D_{eg} \tilde{\rho}_{ee} e^{i(\omega_{ee} - \omega t) t} - \tilde{\rho}_{ee} D_{ge} e^{i(\omega_{ge} + \omega t) t}] + \frac{iE_2}{\hbar} [D_{ee} \tilde{\rho}_{ce} e^{i(\omega_{ee} - \omega t) t} - \tilde{\rho}_{ee} D_{ce} e^{i(\omega_{ce} + \omega t) t}] + \tilde{R}_{ee} \tag{II.1}
$$
The optical coherences become:
\[
\frac{d}{dt} \tilde{\rho}_{\text{ef}} = \frac{i E_{1,0}}{2 \hbar} [D_{\text{ef}} \tilde{\rho}_{\text{ef}} e^{i(\omega_{\text{ef}} - \omega_{\text{f}})t} - \tilde{\rho}_{\text{ef}} D_{\text{ef}} e^{i(\omega_{\text{ef}} - \omega_{\text{f}})t}] + \tilde{R}_{\text{ef}} \tag{II.2}
\]
while the hyperfine coherences are:
\[
\frac{d}{dt} \tilde{\rho}_{\text{ee}} = \frac{i E_{1,0}}{2 \hbar} [D_{\text{ee}} \tilde{\rho}_{\text{ee}} e^{i(\omega_{\text{ee}} - \omega_{\text{e}})t} - \tilde{\rho}_{\text{ee}} D_{\text{ee}} e^{i(\omega_{\text{ee}} - \omega_{\text{e}})t}] + \tilde{R}_{\text{ee}} \tag{II.3}
\]

It is apparent from equations II.4 and II.5 that the optical coherences are coupled not only to the populations but also to the hyperfine coherences.

Appendix III.

Cross product term for broadband excitations. — From equation 7 written in the interaction representation, the perturbation theory gives the « coarse grained » variation of the density matrix as (ref. 9, eq. 5.19):
\[
\tilde{\rho}(t + \Delta t) - \tilde{\rho}(t) = \frac{1}{i \hbar} \int_{t}^{t+\Delta t} dt' \tilde{V}(t'), \tilde{\rho}(t') \int_{t}^{t+\Delta t} dt'' \tilde{V}(t''), \tilde{\rho}(t'') + R. \tag{III.1}
\]
This equation is obtained within the rotating wave approximation. The time increment \( \Delta t \) is limited to values such that \( T_p \gg \Delta t \gg \tau_c \), \( T_p \) and \( \tau_c \) are respectively the pumping time and the coherence time associated to each laser. \( R \) is the contribution of the relaxation.

If we note \( E^{(+)}(t) \) and \( E^{(-)}(t) \) the positive and the negative frequency part of the total electrical field and \( \epsilon_0 \) the polarization vector, we can write:
\[
E(t) = \epsilon_0 E^{(+)}(t) + \epsilon_0^* E^{(-)}(t). \tag{III.2}
\]
In terms of each laser field (supposed parallel and of the same polarization), equation III.2 becomes:
\[
E(t) = \epsilon_0 (E_1^{(+)}(t) + E_2^{(-)}(t)) + \epsilon_0^* (E_1^{(-)}(t) + E_2^{(+)}(t)). \tag{III.3}
\]
This expression is used to evaluate the interaction term and substituted into equation III.1. We recall that \( E_1^{(+)}(t), E_1^{(-)}(t), E_2^{(+)}(t) \) and \( E_2^{(-)}(t) \) are stationary random variables with zero mean value and correlation time (coherence) relatively short. We then take the average value of the resulting equation III.1. The « coarse grained » variation of \( \tilde{\rho}_{\text{e}}(t) \) (sub-matrix associated with the excited state) is given by:
\[
\tilde{\rho}_{\text{e}}(t + \Delta t) - \tilde{\rho}_{\text{e}}(t) = \frac{1}{i \hbar^2} \int_{t}^{t+\Delta t} dt' \int_{0}^{t' - \tau_c} dt'' E_1^{(+)}(t') E_1^{(-)}(t' - \tau) e^{i\omega_{\text{ee}}t''} \times
\]
\[
\times \left[ \{\epsilon_0 \cdot D_1\} (\epsilon_0^* \cdot D_1) \tilde{\rho}_{\text{e}}(t) - (\epsilon_0 \cdot D_1) \tilde{\rho}_{\text{e}}(t) (\epsilon_0^* \cdot D_1) \right]
\]
\[
- E_1^{(+)}(t') E_1^{(-)}(t' - \tau) e^{i\omega_{\text{ee}}t''} (\epsilon_0 \cdot D_1) \tilde{\rho}_{\text{e}}(t) (\epsilon_0^* \cdot D_1)
\]
\[
- E_1^{(+)}(t') E_1^{(-)}(t' - \tau) e^{i\omega_{\text{ee}}t''} (\epsilon_0 \cdot D_1) \tilde{\rho}_{\text{e}}(t) (\epsilon_0^* \cdot D_1)
\]
\[
+ E_2^{(+)}(t') E_2^{(-)}(t' - \tau) e^{i\omega_{\text{ee}}t''} (\epsilon_0 \cdot D_1) \tilde{\rho}_{\text{e}}(t) (\epsilon_0^* \cdot D_1)
\]
+ hermit-conjug.
\[
\text{hermit-conjug.} \tag{III.4}
\]
where \( \rho_{\text{e}} = P_{\text{e}} P_{\text{e}}, D_{\text{e}} = P_{\text{e}} P_{\text{e}} P_{\text{e}}, \rho_{\text{e}}^* = P_{\text{e}} P_{\text{e}} P_{\text{e}}, P_{\text{e}}, P_{\text{e}} \) are the projectors into the levels e, f and g respectively. For sake of simplicity, we have neglected the off resonance terms. The time \( \tau \) is defined by: \( \tau = t'' - t' \).

If the two laser sources are supposed uncorrelated, the second and the third terms under the integral vanish. In this case, resolution of equation III.4 yields, for the population of the excited state, to equation 28. Similar process leads to equations 29 and 30.
References